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Effect of Exchange Interaction on the Low-Temperature Ordering of Gadolinium Trichloride Hexahydrate

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The low-temperature ordered state of Gd^{+++} ions in gadolinium trichloride hexahydrate ($GdCl_3 \cdot 6H_2O$) lattice is investigated taking into account both the dipole-dipole and exchange interactions. It is found that while consideration of the dipole-dipole interaction alone predicts a ferromagnetic low-temperature ordered state, the inclusion of the exchange interaction predicts an antiferromagnetic low-temperature ordered state in agreement with the experimental observations.

I. INTRODUCTION

$GdCl_3 \cdot 6H_2O$ has been observed to undergo antiferromagnetic ordering at low temperatures by Duffy, Lubbers, Van Kempen, Haseda, and Miedema[1] in 1963 and by Wielinga, Lubbers, and Huiskamp[2] in 1967. It is different from the isostructural salts $ErCl_3 \cdot 6H_2O$ and $DyCl_3 \cdot 6H_2O$ in that the latter two undergo ferromagnetic ordering at low temperatures.[3] In Ref. 1 the magnetic properties of $GdCl_3 \cdot 6H_2O$ were investigated by means of adiabatic demagnetization; the zero-field susceptibility was measured in three mutually perpendicular directions, and down to 2 °K the susceptibility was found to follow a Curie-Weiss law with $\theta_x = +0.2\ °K$, $\theta_y = -0.1\ °K$, $\theta_z = -0.3\ °K$. The specific-heat measurements of Ref. 1 yielded the Néel temperature $T_N = 0.182\ °K$. It was conjectured that the antiferromagnetic ordering is caused by the combined effect of crystalline-field splittings and dipolar interactions. The summary of the main points of Ref. 2 is as follows.

(i) The singularity in the heat-capacity curve implies a Néel temperature $T_N = (0.185 \pm 0.001)\ °K$, the broad anomaly lying underneath this singularity being due to Stark splitting of the $^8S_{7/2}$ ground state.

(ii) Zero-field susceptibility measurements yield $\theta_{x'} = +(0.20 \pm 0.15)\ °K$, $\theta_{y'} = -(0.10 \pm 0.10)\ °K$, $\theta_{z'} = -(0.50 \pm 0.20)\ °K$. (For a discussion of the axes see Sec. II.)

(iii) From a comparison of the zero-field susceptibilities in the x', y', and z' directions it follows that the x' axis is the preferred direction of the crystalline field, hence also of antiferromagnetic alignments.

(iv) The doublet $S_{x'} = \pm \frac{7}{2}$ lies lowest as concluded from the width of the heat-capacity singularity, implying a highly anisotropic effective g value ($g_{x'} \gg g_{y'}, g_{z'}$). A theoretical attempt to explain the measurements reported in Ref. 1 was made by Levy[4] who inferred the crystalline field about the Gd^{+++} ions to be described by the potential $V = -0.06[S_{x'}^2 - \frac{1}{3}S(S+1)] + 0.02(S_{y'}^2 - S_{z'}^2)\ \mathrm{cm}^{-1}$, which produces an over-all splitting of about 1 cm^{-1} in the ground state $^8S_{7/2}$ with the doublet $\pm\frac{7}{2}$ lying lowest. Levy also conjectured that the exchange interaction and dipole-dipole coupling give only subordinate contributions to the susceptibility and specific heat. No attempt was made by him to evaluate the low-temperature ordering.

The dissimilarity in the low-temperature ordering of $GdCl_3 \cdot 6H_2O$ (antiferromagnetic) as compared with those of $DyCl_3 \cdot 6H_2O$ and $ErCl_3 \cdot 6H_2O$ (both ferromagnetic) is very curious. The orderings of the latter two can be explained by the consideration of the dipole-dipole interactions alone.[3] Further, it has been conjectured[1-4] that in all these three salts the nondipolar interactions are of subordinate importance. In $GdCl_3 \cdot 6H_2O$, nevertheless, it is suspected that simultaneous consideration of the effect of the crystal field and the exchange interaction on the nominal ground state $^8S_{7/2}$ might modify the prediction based on the consideration of dipole-dipole interaction alone, in favor of the experimentally observed antiferromagnetic ordering.

It is the purpose of this paper to investigate whether the observed low-temperature antiferromagnetic ordering can be explained on the basis of dipole-dipole and exchange interactions that exist between the Gd^{+++} ions in $GdCl_3 \cdot 6H_2O$. The method used is essentially that of Luttinger and Tisza,[5] extended to take into account both the dipole-dipole and the exchange interactions,[6] as well as the presence of two magnetically equivalent ions per unit cell.[7] As far as the low-temperature behavior is concerned, the Gd^{+++} ions will be considered to have an effective spin and an effective g value consistent with the crystal-field splitting[2,3] of the ground state $^8S_{7/2}$. The exchange-interaction con-

TABLE I. Labeling of the 16 ions generating the various sublattices.

Position	Label	Position	Label
$\vec{r}_A$	1	$\vec{r}_B$	9
$\vec{r}_A+\vec{a}$	2	$\vec{r}_B+\vec{a}$	10
$\vec{r}_A+\vec{c}$	3	$\vec{r}_B+\vec{c}$	11
$\vec{r}_A+\vec{a}+\vec{c}$	4	$\vec{r}_B+\vec{a}+\vec{c}$	12
$\vec{r}_A+\vec{b}$	5	$\vec{r}_B+\vec{b}$	13
$\vec{r}_A+\vec{a}+\vec{b}$	6	$\vec{r}_B+\vec{a}+\vec{b}$	14
$\vec{r}_A+\vec{b}+\vec{c}$	7	$\vec{r}_B+\vec{b}+\vec{c}$	15
$\vec{r}_A+\vec{a}+\vec{b}+\vec{c}$	8	$\vec{r}_B+\vec{a}+\vec{b}+\vec{c}$	16

stant will be determined from the measured $\theta_{x'}$, $\theta_{y'}$, $\theta_{z'}$ values and the molecular-field formula.[8]

The crystallographic details of $GdCl_3 \cdot 6H_2O$ crystals are described in Sec. II and a brief outline of the method used for computations is given in Sec. III. Section IV deals with the results of computations and Sec. V with the conclusions.

II. CRYSTAL STRUCTURE OF $GdCl_3 \cdot 6H_2O$

The crystal structure of $GdCl_3 \cdot 6H_2O$ has been determined by Marezio, Plettinger, and Zachariasen[9] from x-ray data. The structure is monoclinic with $\vec{b}$ perpendicular to the $\vec{a}$, $\vec{c}$ plane ($\vec{a}$, $\vec{b}$, $\vec{c}$ are the lattice vectors), the angle between $\vec{a}$ and $\vec{c}$ being 93.65°. There are two Gd^{+++} ions per unit cell (dimensions: $a = 9.651$ Å, $b = 6.525$ Å, $c = 7.923$ Å). The two Gd^{+++} ions occupy magnetically and electrically equivalent sites at $\vec{r}_A = (\frac{1}{4}, 0.1521, \frac{1}{4})$ and at $\vec{r}_B = (\frac{3}{4}, 0.8479, \frac{3}{4})$. For reference purposes, the 16 ions are labeled in Table I. These ions have the following property. If $\Gamma^2 = 2l\vec{a} + 2m\vec{b} + 2n\vec{c}$ (l, m, n being zero, positive, or negative integers), then the 16 sublattices generated by the application of Γ^2 to the positions of ions 1, 2, ..., 16 constitute the entire lattice. The closest distance between Gd atoms is 6.525 Å in the $\vec{b}$ direction; e.g., the nearest neighbors to ion 1 lie on the sublattice generated by the application of Γ^2 to ion 5. It is also noted that the next-nearest neighbors to ion 1 lie on the sublattices generated from ions 13, 14, 15, and 16 at distances 6.55, 6.56, 6.55, and 6.56 Å, respectively.

The principal axes x', y', z' of the magnetic susceptibility for the ordered state, as determined from susceptibility measurement by Wielinga *et al.*,[2] can be related to the directions $\vec{a}$, $\vec{b}$, $\vec{c}$ as: $y' \parallel \vec{b}$; x', z' lie in the $\vec{a}$, $\vec{c}$ plane, the x' axis making an angle of 27° with $\vec{a}$ in a direction away from $\vec{c}$.

III. METHOD OF COMPUTATION

The underlying theory is briefly described as follows. The Hamiltonian describing the interaction between the various ions on the lattice is expressed as

$$\mathcal{H} = \sum_{i<j} \mathcal{H}_{ij} \; ;$$

$$\mathcal{H}_{ij} = v_{ij}\Delta_{ij}\vec{S}_i \cdot \vec{S}_j + \left(\frac{\vec{\mu}_i \cdot \vec{\mu}_j}{r_{ij}^3} - 3\,\frac{(\vec{\mu}_i \cdot \vec{r}_{ij})(\vec{\mu}_j \cdot \vec{r}_{ij})}{r_{ij}^5} \right) , \tag{1}$$

where the summation is over all the ions in the crystal. The first term of $\mathcal{H}_{ij}$ represents the exchange interactions and the second term the dipole-dipole interactions. Here $\vec{r}_{ij}$ is the vector that joins the ion i to ion j and $\vec{\mu}_i = \mu_B \overleftrightarrow{g} \cdot \vec{S}_i$, where μ_B is the Bohr magneton, $\overleftrightarrow{g}$ is the g tensor, and $\vec{S}_i$ is the effective spin. (A coordinate system in which the g tensor is diagonal is chosen.) In Eq. (1) $\Delta_{ij} = 1$, when i and j are nearest and next-nearest neighbors, 0 otherwise, and v_{ij} is the exchange-interaction constant between ions i and j. An isotropic exchange has been assumed for the present calculations.

If a set of wave functions which are the direct products of 16 one-spin wave functions is used, the expectation value of the energy per ion in the lattice can be written

$$E = \sum_{i,j=1}^{16} \; \sum_{\alpha,\beta=x,y,z} A_{ij}^{\alpha\beta} \xi_i^\alpha \xi_j^\beta \; , \tag{2}$$

where

$$A_{ij}^{\alpha\beta} = \sum_{l \in \{j\}} J_{il}^{\alpha\beta} \quad \text{for } i \neq j \; ,$$

$$A_{jj} = \sum_{\substack{l \in \{j\} \\ \neq j}} J_{jl}^{\alpha\beta} \; ; \tag{3}$$

$$J_{ij}^{\alpha\beta} = \frac{S^2}{2} \left[\frac{\mu_B^2 g^\alpha g^\beta}{r_{ij}^3} \left(\delta_{\alpha\beta} - \frac{3 r_{ij}^\alpha r_{ij}^\beta}{r_{ij}^2} \right) + v_{ij}\Delta_{ij}\delta_{\alpha\beta} \right] . \tag{4}$$

(The Greek indices run over x', y', z' and the Latin indices over 1, 2, ..., 16.)

In Eq. (2), ξ_i^α represents the expectation value of the operator $\hat{S}_i^\alpha / S$. In Eq. (3) $\{j\}$ denotes the set of lattice sites generated by the application of Γ^2 to ion j.

The two conditions, related to the normalization of the spin components, are as follows:

$$\text{"strong" condition:} \quad \vec{\xi}_i \cdot \vec{\xi}_i = 1 \; , \tag{5}$$

$$\text{"weak" condition:} \quad \sum_{i=1}^{16} \vec{\xi}_i \cdot \vec{\xi}_i = 16 \; . \tag{6}$$

The task is to find the lowest value of E consistent with the "strong" condition. On the other hand, consistent with the "weak" condition the lowest value of E is the lowest eigenvalue of the 48×48 matrix with the components $A_{ij}^{\alpha\beta}$ given by Eq. (3) above. The problem is solved if the resulting eigenvectors of A also satisfy the "strong"

TABLE II. Lattice sums for $GdCl_3 \cdot 6H_2O$. The variables R, X, Y, and Z are the displacements and its components from an origin situated at a given ion in sublattice 1 to each ion in the various sublattices. The unit of length is $0.2a$; $a = 1.0085$ Å. Note that $A \equiv \sum(3Z^2 - R^2)/R^5$, $B \equiv \sum(X^2 - Y^2)/R^5$, $C \equiv \sum XY/R^5$, $D \equiv \sum XZ/R^5$, $E \equiv \sum YZ/R^5$.

Sublattice summed	A	B	C	D	E
1	8.7869	−2.3514	−0.1820	0	0
2	−15.3394	17.7224	0.1733	0	0
3	−25.9104	−33.3059	2.1117	0	0
4	−11.5812	3.3577	0.9974	0	0
5	115.9501	−2.1969	−0.1589	0	0
6	−2.4163	11.4982	0.1557	0	0
7	6.5493	−16.2696	−1.0226	0	0
8	−5.7295	2.8361	0.7190	0	0
9	5.6669	1.4466	5.8197	4.5239	3.7607
10	2.9635	2.0880	−5.1048	−3.8801	2.8358
11	2.9635	2.0880	−5.1048	3.8801	−2.8358
12	5.6669	1.4466	5.8197	−4.5239	−3.7607
13	−20.5212	3.5883	13.2731	−6.3328	−5.3284
14	−18.6800	4.8602	−10.7874	5.1377	−3.8078
15	−18.6800	4.8602	−10.7874	−5.1377	3.8078
16	−20.5220	3.5883	13.2731	6.3328	5.3284

condition. That such a situation does indeed hold true for a lattice with two magnetically equivalent ions per unit cell has been proven in Ref. 7 based on the Luttinger-Tisza method,[5] further extended by Niemeyer to include exchange interactions.[6] The details are briefly described below.

Let P_t belong to the set of permutations listed in Ref. 7, such that

$$A^{\alpha\beta}_{P_t i, P_t j} = A^{\alpha\beta}_{ij} .$$

Then the eigenvectors of A are given as

$$\eta^{\nu}_{j}(k, \gamma) = q_j(k)\Phi^{\nu}(\gamma) \tag{8}$$

($k = 1, 2, \ldots, 16$; $\gamma = x', y', z'$). The $\vec{q}(k)$ are the eigenvectors of P_t (listed in Ref. 7). The $\Phi^{\nu}(\gamma)$ are the solutions of the eigenvalue equation

$$L^{\mu\nu}_{k}\Phi^{\nu}(\gamma) = \lambda^{l}_{k}\Phi^{\mu}(\gamma) ,$$

where

$$L^{\mu\nu}_{k} = \sum_{j} A^{\mu\nu}_{ij}\epsilon_{P(i,j)}(k) . \tag{9}$$

(The summation on the right-hand side of Eq. (9) is independent of i.[6]) In Eq. (9), $\epsilon_{P(i,j)}(k)$ are the eigenvalues of the permutation operators $P(i,j)$, containing the cycle (i,j) with respect to the eigenvector $\vec{q}(k)$ ($\epsilon_{P(i,j)}$ are listed in Ref. 7). From the form of the $\vec{q}(k)$'s it is readily seen that the eigenvectors $\eta(1,\gamma)$ and $\eta(2,\delta)$ correspond to ferromagnetic and antiferromagnetic arrangements of spins, respectively, while the $\eta(i,\epsilon)$; $i = 3, 4, \ldots, 16$, correspond to layered antiferromagnetic arrangements of spins.

IV. DETAILS OF COMPUTATION AND RESULTS

The effect of the crystal field on the ground-state splitting of Gd^{+++} in $GdCl_3 \cdot 6H_2O$ (of order $1\ cm^{-1}$) must be duly taken into account. This splitting causes the $S_{x'} = \pm\frac{7}{2}$ doublet to lie lowest, and in the temperature range within which the magnetic ordering takes place only this doublet is significantly populated. The ground state of Gd^{+++} at low temperatures can thus be described by an effective spin-$\frac{1}{2}$ with a highly anisotropic g factor ($g_{x'} \gg g_{y'}, g_{z'}$). It is reasonable to assume that for this doublet the splitting in a magnetic field would be described by a $g_{x'} = 7 \times 1.999 = 13.993$ [1.999 is the $g(= g_{x'} = g_{y'} = g_{z'})$ factor at higher temperatures where all the sublevels of spin-$\frac{7}{2}$ are occupied[10]].

As for the exchange interaction, the molecular-field-theory formula $\theta_{\text{powder}} = -zS(S+1)v/3k$ is used,[8] where $z(=2)$ is the number of nearest neighbors to a Gd atom, v and k are the exchange and Boltzmann constants, respectively. We assume $\theta_{\text{powder}} = \theta_{x'} + \theta_{y'} + \theta_{z'}$ and use the values measured by Wielinga *et al.*[2] This yields $v = 0.800$ °K.

Since $g_{x'} = g_{y'} = 0$ many of the elements of the matrix A are zero. In particular, for use in Eq. (3), one has

$$J^{x'\alpha'}_{ij} = J^{\alpha'x'}_{ij} = \frac{S^2}{2}\left[\frac{\mu_B^2 g_{x'}^2}{r_{ij}^3}\left(1 - \frac{3x_{ij}'^2}{r_{ij}^2}\right) + v_{ij}\Delta_{ij}\right]\delta_{\alpha'x'}$$

and

$$J^{\alpha\beta}_{ij} = \tfrac{1}{2}S^2 v_{ij}\delta_{\alpha\beta} \quad \text{for } \alpha, \beta = x', y' . \tag{10}$$

The eigenvalues of A as seen from Eq. (10) are its diagonal elements since the off-diagonal elements of A are zero. The short-range exchange contribution from ions other than the nearest and next-nearest neighbors are neglected. As far as the long-range dipole-dipole interactions are concerned, all ions within a sphere of radius 500 Å are taken into account. The relevant lattice sums are presented in Table II. A coordinate system X, Y, Z has been used with $X \parallel \vec{a}$, $Z \parallel \vec{b}$. The only lattice sums $\sum_j (r_{1j}^2 - 3x_{1j}'^2)/r_{ij}^5$ required for the present calculations can be obtained in terms of the lattice sums in the X, Y, Z system as follows:

$$\sum_j \frac{r_{1j}^2 - 3x_{1j}'^2}{r_{1j}^5} = \frac{1}{2}\sum_j \frac{3Z_{1j}^2 - R_{1j}^2}{R_{1j}^5} + \tfrac{3}{2}\cos(54°)\sum_j \frac{X_{1j}^2 - Y_{1j}^2}{R_{1j}^5} + 3\sin(54°)\sum_j \frac{X_{1j}Y_{1j}}{R_{1j}^5}$$

(note: $r_{ij}^2 = R_{ij}^2 = X_{ij}^2 + Y_{ij}^2 + Z_{ij}^2 = x_{ij}'^2 + y_{ij}'^2 + z_{ij}'^2$).

It should be noted further that if the summations were performed over a sphere of radius larger than 500 Å, the results would be negligibly different from those given in Table II, as checked by repeating the sums over a sphere of radius 600 Å.

Since the lattice sums are evaluated over a sphere, the demagnetization factors (D_F)[7] for the eigenvalues $E_1^{x'}$, $E_1^{y'}$, $E_1^{z'}$, which correspond to

TABLE III. Eigenvalues for $GdCl_3 \cdot 6H_2O$. The demagnetization factors should be added to the eigenvalues E_1, E_2, and E_3 corresponding to the ferromagnetic orderings. The three eigenvalues corresponding to the eigenvector $\vec{q}(k)$ are denoted with the numbers $(3k-2)$, $(3k-1)$, and $3k$, respectively. In each case, $E_i = E_i^{dd} + E_i^{ex}$. Note: v_{nn} and v_{nnn} denote the nearest- and the next-nearest-neighbor exchange-interaction constants, respectively.

Dipole-dipole contributions in °K (E_i^{dd})	
$E_1^{dd} = 0.022333$	$E_{25}^{dd} = E_{28}^{dd} = 0.252368$
$E_4^{dd} = 0.156785$	$E_{31}^{dd} = E_{34}^{dd} = -0.109385$
$E_7^{dd} = -0.187338$	$E_{37}^{dd} = 0.440689$
$E_{10}^{dd} = 0.036950$	$E_{40}^{dd} = -0.206397$
$E_{13}^{dd} = E_{16}^{dd} = 0.028324$	$E_{43}^{dd} = -0.079443$
$E_{19}^{dd} = -0.044741$	$E_{46}^{dd} = -0.079441$
$E_{22}^{dd} = -0.222822$	$E_{3k-1}^{dd} = E_{3k}^{dd} = 0;\ k = 1, 2, \ldots, 16$
Exchange contributions (E_i^{ex})	
$E_{3k}^{ex} = E_{3k-1}^{ex} = E_{3k-2}^{ex};\ k = 1, 2, \ldots, 16$	
$E_3^{ex} = -E_{21}^{ex} = \frac{1}{8}(2v_{nn} + 8v_{nnn})$	
$E_6^{ex} = -E_{24}^{ex} = \frac{1}{8}(2v_{nn} - 8v_{nnn})$	
$E_{3k}^{ex} = -\frac{2}{8}v_{nn};\ k = 3, 4, 11, 12, 15, 16$	
$E_{3k}^{ex} = \frac{2}{8}v_{nn};\ k = 5, 6, 9, 10, 13, 14$	

the ferromagnetic arrangement of spins must be included. In general, $D_F^\alpha = -\frac{2}{3}\pi\mu_B^2 g^{\alpha 2} S^2 n_0$, where n_0 is the number of ions per cm^3. The demagnetization factors relevant to the present problem are $D_F^{x'} = -0.25708$ °K and $D_F^{y'} = D_F^{z'} = 0$.

The eigenvalues of A are listed in Table III. In particular, the two lowest-lying eigenvalues are (assuming $v_{nnn} \ll v_{nn}$ and that the effect of v_{nnn} is negligible insofar as the two lowest-lying eigenvalues are concerned. In any case, even if v_{nnn} were not neglected, an antiferromagnetic ordering would still correspond to the lowest-lying eigenvalue, because of the particular exchange contribution to $E_1^{x'}$ and $E_8^{x'}$)

ferromagnetic

$$E_1^{x'} = E_1^{x'(dd)} + E_1^{x'(ex)} = -0.03475\ °K\ ,$$
$$E_1^{x'(dd)} = -0.23475\ °K\ , \quad E_1^{x'(ex)} = 0.2 + v_{nnn}\ ; \tag{11}$$

layered antiferromagnetic

$$E_8^{x'} = E_8^{x'(dd)} + E_8^{x'(ex)} = -0.42282\ °K\ ,$$
$$E_8^{x'(dd)} = -0.22282\ °K\ , \quad E_8^{x'(ex)} = -0.2 + v_{nnn}\ . \tag{12}$$

(In both the above cases the axis of alignment of spins is the x' axis.)

Here v_{nnn} represents the next-nearest-neighbor exchange constant (its value is irrelevent for the present considerations since both $E_1^{x'}$ and $E_8^{x'}$ receive equal exchange contributions from the next-nearest neighbors). From (11) and (12) it is clearly seen that a layered antiferromagnetic configuration lies lowest (as long as $v > 0.02386$ °K), the next lowest eigenvalue corresponding to a ferromagnetic ordering, i.e., $E_8^{x'} < E_1^{x'}$. It should also be noted that if exchange interactions were *not* considered, a lowest eigenvalue corresponding to ferromagnetic ordering would have been found.

V. CONCLUSIONS

The present investigations indicate that the exchange interaction plays an important role in affecting the low-temperature ordering of $GdCl_3 \cdot 6H_2O$. On the basis of dipole-dipole interaction alone a ferromagentic ordering of spins would have been affected, whereas taking the exchange interaction into account predicts the low-temperature ordering to be antiferromagnetic in agreement with the experimental observations. The crystal-field splitting of the $^8S_{7/2}$ ground state of Gd^{+++} ions in $GdCl_3 \cdot 6H_2O$ has been accommodated by assigning an effective spin-$\frac{1}{2}$ to the ions with appropriate values for $g_{x'}$, $g_{y'}$, $g_{z'}$.

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